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[54]名　稱：有機電激發光元件圖文顯示的製作方法

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[57]申請專利範圍：

1.一種有機電激發光元件顯示圖文的製作方法，用以在一透光導電玻璃基板上形成該有機電激發元件，該方法包括下列步驟：
在該透光導電玻璃基板上，形成一有機導電層；
在該有機導電層中，形成一顯示圖案、文字的區域；
在該有機導電層上，形成一有機發光層；
在該有機發光層上，形成一金屬電極；以及
形成該有機電激發光元件。

2.如申請專利範圍第1項所述之製作方法，其中形成該有機導電層之方法包括旋轉塗佈法。

3.如申請專利範圍第1項所述之製作方法，其中該有機導電層係選自於由聚乙炔、聚苯胺、聚吩、聚對位苯基乙烯、聚對位苯、聚乙烯吩、聚硫苯和聚氧化二甲苯，以及以上各種化合物的衍生物、自身摻雜衍生物與共聚物衍生物所組成之族群中的材料。

4.如申請專利範圍第1項所述之製作方法，其中該有機導電層之材料係由一有機導電材料與一傳統高分子材料所構成的摻合體。

5.如申請專利範圍第4項所述之製作方法，其中該有機導電材料係選自於由聚乙炔、聚苯胺、聚吩、聚對位苯基乙烯、聚對位苯、聚乙烯吩、聚硫苯和聚氧化二甲苯，以及以上各種化合物的衍生物、自身摻雜衍生物與共聚物衍生物所組成之族群中的材料。

6.如申請專利範圍第4項所述之製作方法，其中該傳統高分子材料係選自於由聚

甲基丙烯酸甲酯、聚乙烯醇、聚環氧乙烷、聚對苯二甲酸乙二酯及聚丙烯醯胺所組成之族群中的材料。

7.如申請專利範圍第 4 項所述之製作方法，其中該傳統高分子材料之摻合比例約為 1%～ 99%。

8.如申請專利範圍第 1 項所述之製作方法，其中形成該顯示圖案、文字的區域之方法包括使用紫外光照射。

9.如申請專利範圍第 1 項所述之製作方法，其中形成該顯示圖案、文字的區域之方法包括使用電子束照射。

10.如申請專利範圍第 1 項所述之製作方法，其中形成該顯示圖案、文字的區域之方法包括使用雷射掃瞄。

11.如申請專利範圍第 1 項所述之製作方法，其中形成該有機發光層之方法包括旋轉塗佈法。

12.如申請專利範圍第 1 項所述之製作方法，其中形成該有機發光層之方法包括蒸鍍法。

13.如申請專利範圍第 1 項所述之製作方法，其中形成該金屬電極之方法包括蒸鍍法。

圖式簡單說明：

第一圖 A 至第一圖 C 繪示習知一種以陰極分隔柱製作有機電激發光顯示元件之製作流程圖。

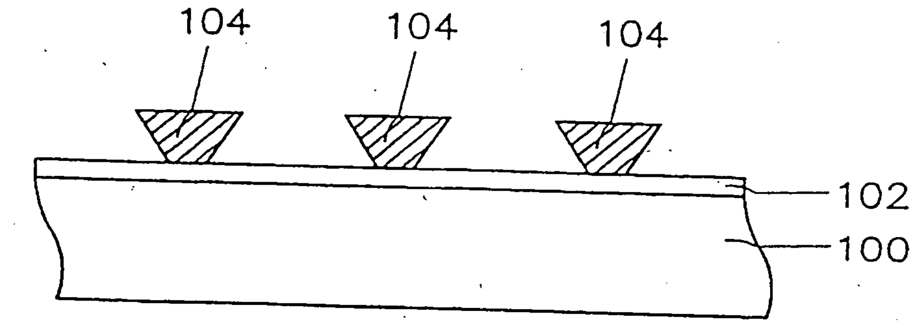
第二圖 A 至第二圖 F 繪示習知一種以光阻 -曝光程序對金屬電極定型化製作有機電激發光顯示元件之製作流程圖。

第三圖 A 至第三圖 C 繪示習知一種將發光層定型化之有機電激發光顯示元件的製作流程圖。

第四圖繪示習知一種具有定型化發光層之有機電激發光顯示元件的結構剖面圖。

第五圖 A 至第五圖 E 繪示依照本發明之一較佳實施例，一種應用於固定式顯示圖案、文字之有機電激發光元件的製作流程圖。

第一圖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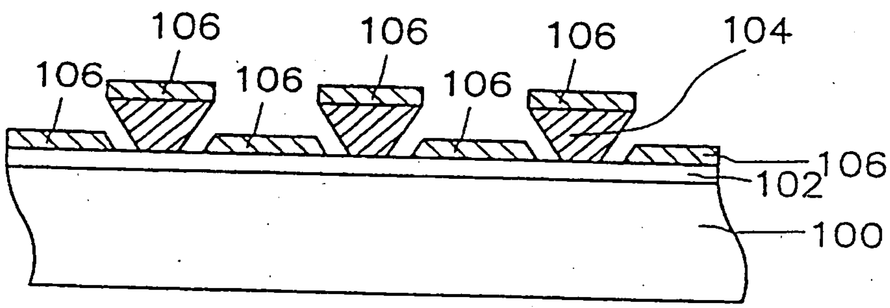
102
100

A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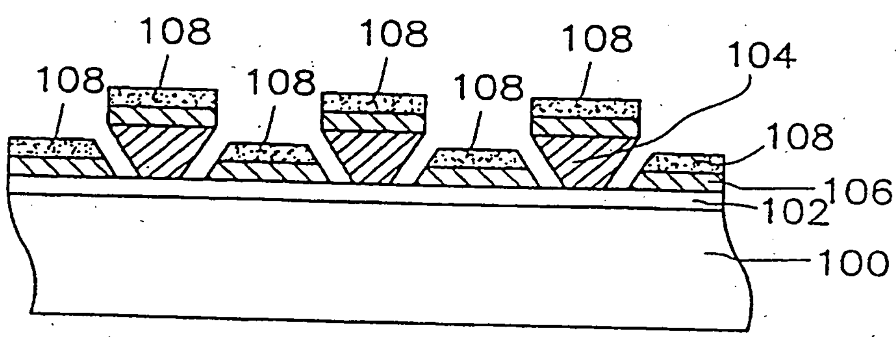
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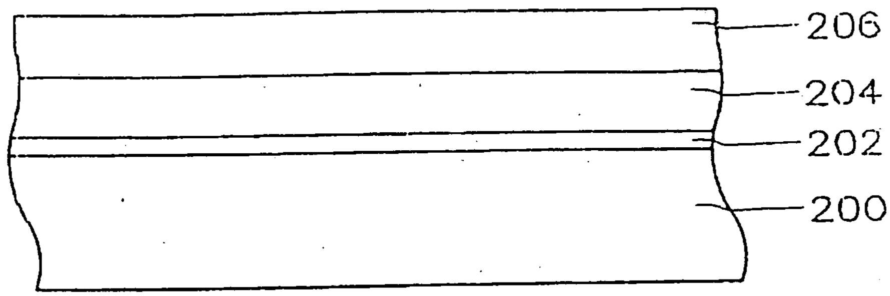
C

第二圖



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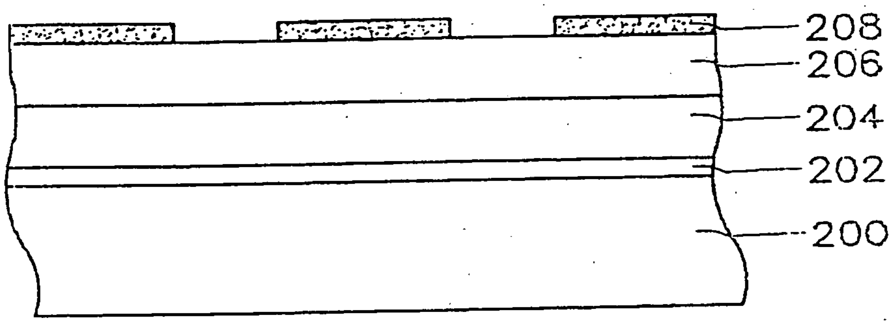

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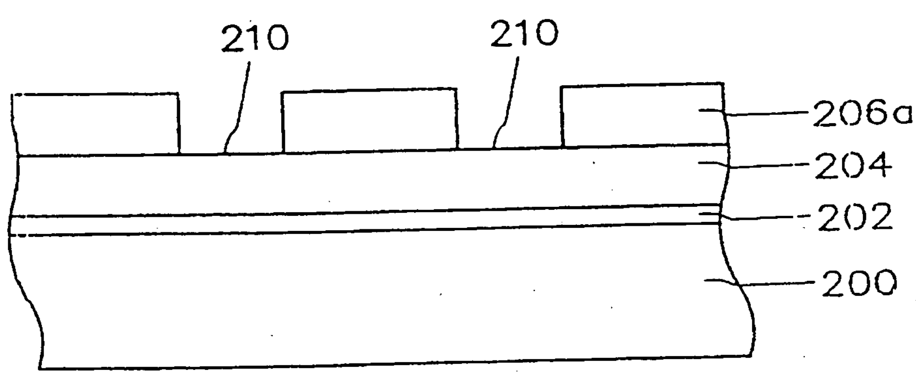

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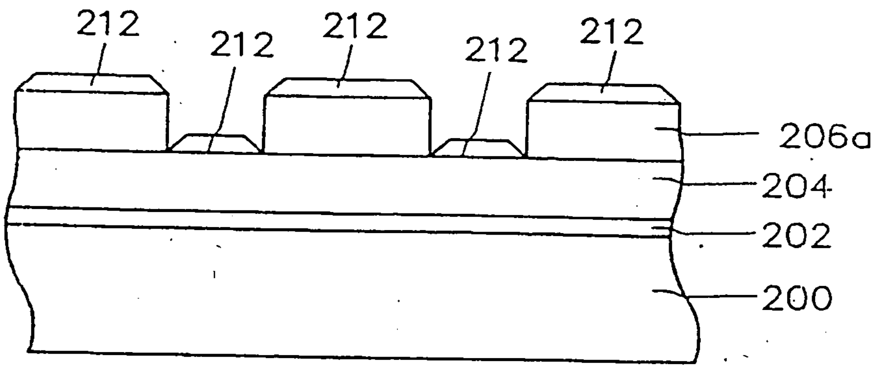
第二圖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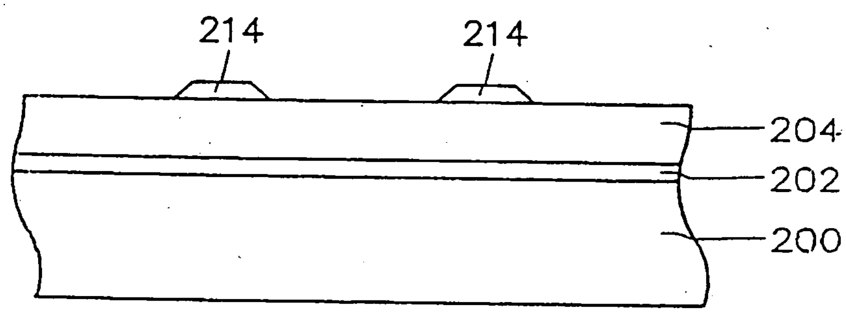
D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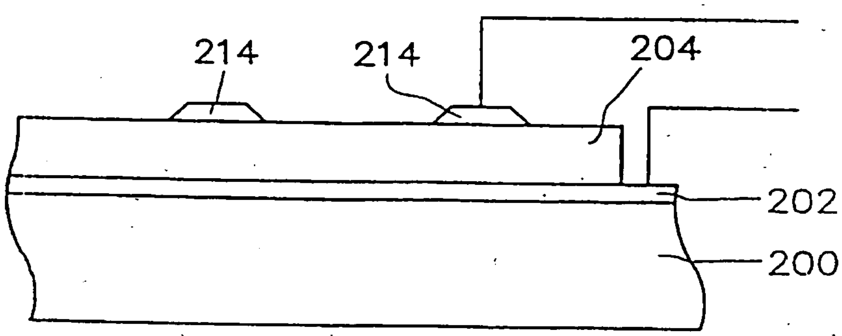
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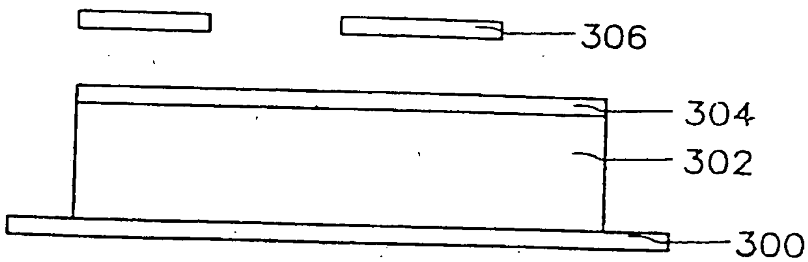
F

第三圖



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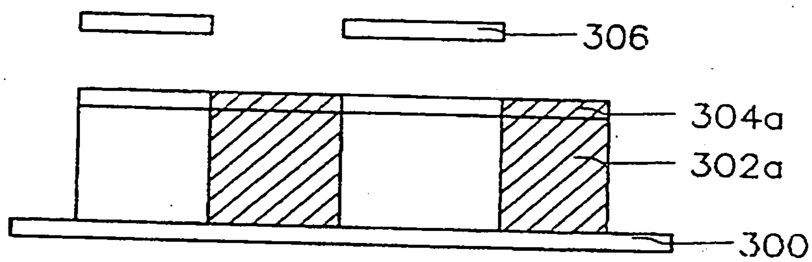

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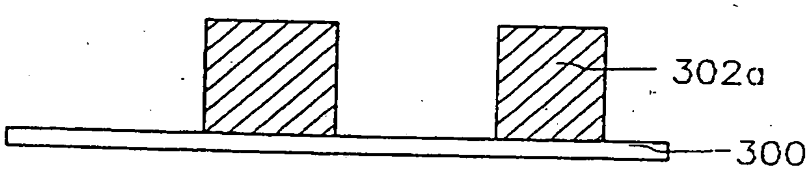

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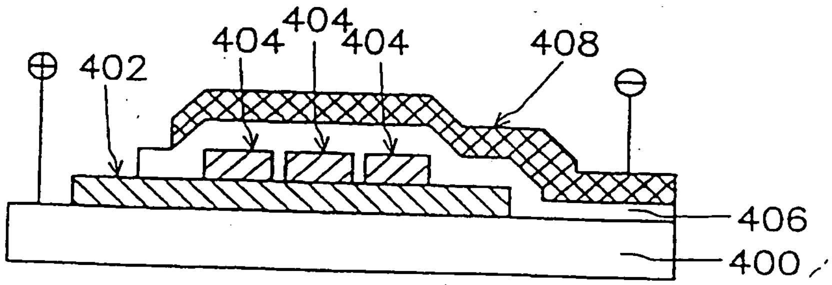
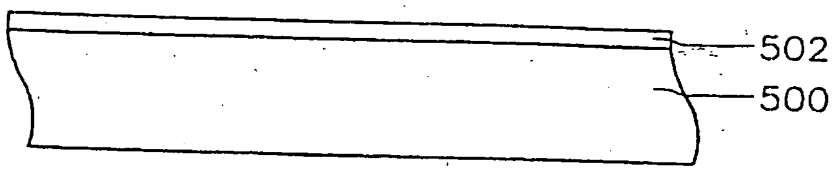


第五圖



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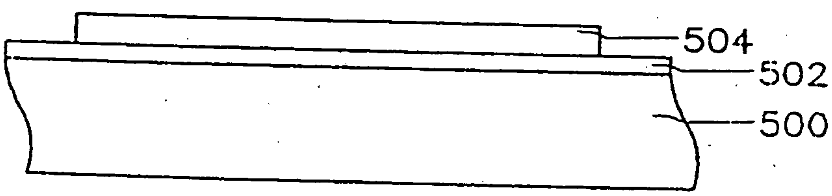

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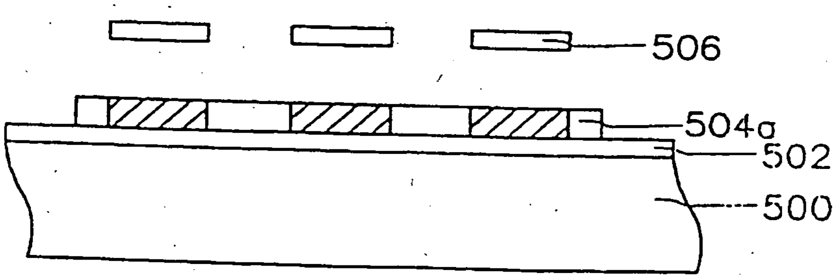

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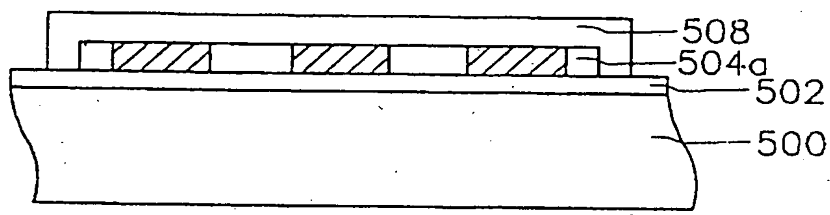
第五圖



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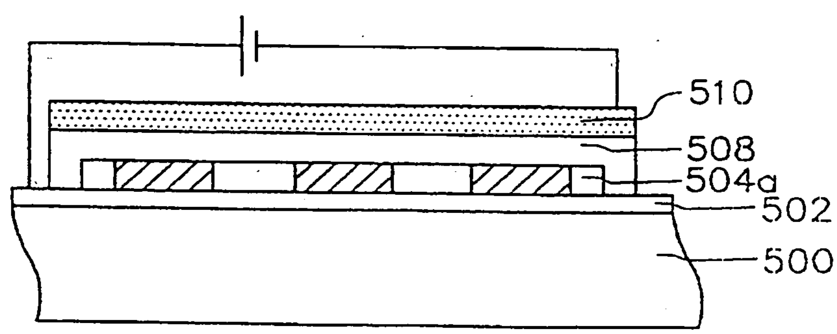

E

TW406518

Method for manufacturing an organic electro-luminescent device for picture/character displaying

A method for manufacturing a static-type organic electro-luminescent device for picture/character displaying, comprising the steps of: providing a transparent conductive glass substrate, forming an organic conductive layer on the substrate, patterning the conductive layer by optical-chemical reaction, forming regions for displaying picture/character in said organic conductive layer, then sequentially forming an organic light emitting layer, metal electrode and metal protection layer on said patterned organic conductive layer as to complete the manufacturing of the device. The organic electro-luminescent device may display desired picture or character when the patterned organic conductive layer is applied with proper bias. As the organic light emitting layer is formed by vaporization or spin-coating, it can be suitably applied to a polymer organic electro-luminescent device formed by solution spin-coating process and a small molecule organic electro-luminescent device form by evaporation process.

J00087I2TW01

[Field of the Invention]

The invention relates to a manufacturing method for a static organic electro-luminescent (OEL) device for displaying pictures and characters, and more particularly, to a method for manufacturing a static-type organic electro-luminescent device for displaying pictures and characters by using pattern organic conductive films.

[Description of Related Art]

It has been over 30 years from the development of organic electro-luminescence in 1960s. In 1963, a research report firstly announced that a single crystal organic compound illuminates when a high voltage of 400 voltages is applied. However, the intensity of luminescence was far below the required intensity for practical usage.

In 1987, Kodak USA announced an electro-luminescent device of small molecule (Applied Physics Letter, Volume 51, pp.914, 1987). In 1990, the Cambridge University successfully applied polymer material onto the electro-luminescent device (Nature, Volume 347, pp.539, 1990). Both of these technologies have become the foundation for the practicality of the electro-luminescent device. Since then, the continuous research and development by numerous academic and industrial research studies was triggered.

The electro-luminescent device has the feature of self-luminescence, wide view angle (up to 160°), high responsibility, low driving voltage, and full color etc and has been recognized to be a flat panel display technology of next century. The development on the organic electro-luminescent device has almost reached a practical stage, it promises to be applied to next-generation flat panel color displays, including small-size display panels, outdoor display panels, and monitors, etc.

The current development of organic electro-luminescent device stresses on device and material configuration thereof, the displaying technology for picture and characters is lesser-noticed. The displaying mode to picture and character for organic electro-luminescent device is roughly classified into static-type displaying mode and dynamic-type displaying mode. The dynamic displaying mode is used to provide an image of moving pictures or characters as to be applied to such as digital display, dynamic advertisement board, monitor display, etc. But, such displaying mode requires a dot matrix element for displaying each pixel and a complicated control circuit system such that the cost and the technical requirement are high. Static displaying mode can merely display a fixed image message of pictures or characters and used as for example slogan indicator, signs, automobile panel, fixed-type advertisement board. However, it merely requires a suitable area of flat display device and a simpler operating circuit, it thus costs a lesser technical requirement and cost.

Therefore, both of these two display modes have different market shares.

There is a demand to improve a manufacturing method using spin-coating process to manufacture a polymer electro-luminescent device for displaying pictures and characters. The manufacturing technology for organic electro-luminescent device of static type is described as followed:

Referring Figs. 1A-1C, a manufacturing flow for organic electro-luminescent device with cathode separator for displaying picture and character is shown. Such a method was proposed by Princeton University (Applied Physics Letter, Volume 77, pp3197, 1997) and Pioneer Corporation (Jpn. J. Appl. Phys., Vol.36, p1555 (1997)) and used a metal electrode pixelization to form an organic electro-luminescent device of small molecule.

Referring to Fig. 1A, an indium tin oxide (ITO) glass substrate consisted of a glass substrate 100 and an ITO layer 102 is provided. Then, T-shaped cathode separating poles 104 are formed on the ITO glass substrate for separating the metal electrodes later formed. In Fig. 1B, organic material is vaporized onto the ITO layer 102s and T-shaped cathode separating poles 104 as to form organic light emitting layer 106. Referring Fig. 1C, metal electrodes 108 are formed on organic light emitting layer 106 by vaporization thereby completing the manufacturing of displaying device. Such a manufacturing method is complicated and is not suitable to a displaying device of polymer using solution spin-coating.

Referring to Figs. 2A-2F, a manufacturing flow for patterning metal electrode of organic electro-luminescent display device by photoresist-exposure process is shown. This technology is proposed by Lidzey (Synth. Met., Vol.82, p141,1996) for manufacturing metal electrodes of micro-pattern in a device by photoresist-exposure process in organic electro-luminescent device of polymer.

As shown in Fig. 2A, an indium tin oxide glass substrate consisted of a glass substrate 200 and indium tin oxide layer 202 is provided. An organic light emitting layer 204 and photoresist 206 are sequentially formed on the substrate, wherein the material of organic light emitting layer is poly(2.5-dialkoxy-p-phenylene-vinylene) (PDAOPV). Referring 2B, photoresit 206 with contact mask 208 is exposed by ultraviolet light. After exposure, a patterned photoresist 206A and opening 210 is formed as shown in Fig. 2C. As shown in Fig. 2D, a metal layer 212 is formed on pattered photoresist 206 and a bottom of the opening 210 with vaporization. The patterned photoresist 206a and portion of metal layer 212 is removed by acetone and metal electrodes 214 are formed on the organic light emitting layer 204 as shown in Fig. 2E. The metal electrodes are then electrically connected with exterior as shown in Fig. 2F. The initial electric field of the device manufactured by this process is increased and the illumination efficiency thereof is reduced to 35% in comparison

with that of the mask-pattern type device. This is resulted from the processes of photoresist coating, baking, ultraviolet exposure, development and photoresist removing, thus, the quantum efficiency of phosphor of light emitting material is reduced.

Referring Figs. 3A-3C, a manufacturing flow chart for patterning a light emitting layer of an organic electro-luminescent device is shown. The process is developed by HP corp. and Rochester university (Advanced Materials, Vo. 9, p392, 1997) which patterns the organic light emitting layer by using a difference between poly(phenylene vinylene)(PPV) and its precursor with photolithographic process as to produce polymer device.

In Fig. 3A, an indium tin oxide glass substrate coated with organic film is provided, wherein a poly(phenylene vinylene) layer 302 and a mixture layer 304 of polybutadiene (PBD) and polystyrene (PS) is coated on said indium tin oxide layer 300. Then, the mask 306 is exposed such that the organic film generates a photogenerated acid catalyzes elimination. After exposure, the mask pattern of the mask is transferred to organic film and a layer 302a of patterned poly(phenylene vinylene) and a mixture layer 304a of patterned polybutadiene and polystyrene are formed as shown in Fig. 3B. After further exposure, a light emitting layer of patterned poly(phenylene vinylene) 302a is shown in Fig. 3C. From the above–mentioned steps, a polymer device of ITO/pattern PPV/PBD:PS mixture layer/Ca is obtained. Referring Fig. 4, a cross sectional view of organic electro-luminescent device with patterned light emitting layer is shown. The device is consisted of glass substrate 400, ITO layer 402, patterned organic electro-luminescent layer 404, electron transport layer 406, and metal electrodes 408, etc.

The pattern feature of such process is about to 125 μm, but the current leakage is large such that the quantum efficiency thereof is reduced to 0.01% from 0.25% of normal processes. Such process may merely be applied to few phosphor with precursor and the illumination efficiency of the material is reduced during exposure. The clearness of material surface is influenced in development.

In addition to said process, a method provided by Yang Yang using an ink-jet printer, instead of a spin-coater, to coat polymer is able to reduce the waste of polymer in the fabrication process (Science, Volume 279, p.p. 1135, 1998). This method has the advantages of saving polymer material, making any desired patterns, characters with the droplet of 30 μm, and full-color displaying device. However, this method still has some problems to be overcome, such as the transportation of ITO glass, the utilization of solution and inkjet head jam.

As the organic electro-luminescent device has the advantages of high responsibility, low driving voltage, full-color display, it has been recognized to be a

flat panel display technology of next century. The current development of organic electro-luminescent device stresses on device and material configuration thereof, the displaying technology for picture and characters is lesser-noticed. The disadvantages and problems of existing displaying technologies are listed in table 1.

Table 1 Display Technologies Analysis of Organic EL

Item	Method	Developer	Disadvantage
1	Cathode spacer	Pioneer; Princeton Univ.	a. High technical requirement and complicated, not suitable for static picture display b. not suitable polymer device with solution coating
2	ITO glass pattern		a. ITO glass patterning requires placement of resist, exposure, development, etching, removal of resist. b. un-evenness of illumination of pictures is generated by gradient distribution of electric field in high resolution, low line width
3	Light emitting layer patterning	HP & Rochester Univ.	a. Light-emitting layer patterning requires placement of resist, exposure, development, etching, removal of resist b. These processed will reduce the illumination efficiency of the surface and thus the efficiency of the device
4	Patterning metal electrode with photoresist	D.G. Lidzy et. al (Sheffield Univ.)	a. Metal electrodes patterning requires placement of resist, exposure, development, etching, removal of resist b. These processes will reduce the clarity of the surface and thus the efficient of the device
5	Patterning metal electrode with mask		a. A simple procedure but limited by metal mask, the resolution is low b. The problem of electric field distribution is induced when the resolution is increased
6	Ink-jet printer	Yang Yang (UCLA)	a. Reduce the cost of polymer material for manufacturing any picture display for full color display

			b. A new procedure with the difficulties of transferring of ITO glass, solution utilization and inkjet head jam

Accordingly, the main objective of the present invention is to provide a method for forming an organic electro-luminescent device of static display type. In particular, the present invention provides a manufacturing method which is suitable for either polymer organic electro-luminescent device by using solution coating process or small molecule organic electro-luminescent device by using evaporation.

In accordance with the foregoing objective of the present invention, the method of the invention for forming organic electro-luminescent device for displaying pictures and characters, comprising the steps of: providing a transparent conductive glass substrate, forming an organic conductive layer on the substrate after cleaning the surface of the substrate, patterning the organic conductive layer, forming regions for displaying picture/character in said organic conductive layer, then forming an organic light emitting layer on said patterned organic conductive layer by coating or evaporation material of proper hole and electron transport layers, and then sequentially forming proper metal electrode and protection layer on the surface of said organic light emitting layer. The organic electro-luminescent device may display desired pictures or characters when a proper bias is applied by having the transparent conductive glass substrate as positive electrode while the metal electrode as negative electrode.

[Brief Description of Drawings]

The invention can be more fully understood by reading the following detailed description of the preferred embodiments, with reference made to the accompanying drawings, wherein:

Figs. 1A-1C are schematic cross-sectional views showing a manufacturing flow for a conventional organic electro-luminescent device formed with cathode spacer;

Figs. 2A-2F are schematic cross-sectional views showing a manufacturing flow for patterning metal electrodes with a photo-exposure process of a conventional organic electro-luminescent device;

Figs. 3A-3C are schematic cross-sectional views showing a manufacturing flow for patterning a light emitting layer of a conventional organic electro-luminescent device;

Fig. 4 is a cross-sectional view of a conventional organic electro-luminescent device with patterned light emitting layer; and

Figs. 5A-5E are manufacturing flows for an organic electro-luminescent device

applied to static displaying according to an embodiment of this invention.

[List of major elements]

100, 200, 300, 400, 500	glass substrate
102, 202, 302, 402	ITO layer
104	cathode spacer
106, 204, 508	organic light emitting layer
108, 214, 408, 510	metal electrode
206	photoresist
206a	patterned photoresist
208, 306, 506	photomask
210	opening
212	metal layer
302	poly(phenylene vinylene) layer
302a	patterned poly(phenylene vinylene) layer
304	mixed layer of polybutadiene & polystyrene
304a	patterned mixed layer of polybutandiene & polystyrene
404	patterned organic electro-luminescent layer
406	electron transport layer
502	transparent conductive layer
504	organic conductive layer
504a	patterned organic conductive layer

[Description of Embodiment]

Referring to Figs. 5A-5E, a manufacturing flow for organic electro-luminescent device for static displaying in accordance with an embodiment of this invention is shown. As shown in Fig. 5A, a transparent conductive glass substrate is provided, it is consisted of glass substrate 500 and transparent conductive layer (TCL) 502, which is for example indium tin oxide (ITO). The surface of the substrate is cleaned for further processes.

Referring to Fig. 5B, an organic conductive layer 504 is formed on the surface of transparent conductive layer 502 by, for example, spin coating. The organic conductive layer 504 is made from an organic conductive material, for example, polyacetylene (PA), polyaniline (PAn), polythiophene (PT), poly(phenylene vinylene) (PPV), poly(paraphenylene) (PPP), polythiophene vinylene (PTV), poly(phenylene sulfide)(PPS), poly(phenylene oxide) (PPO) and their derivatives, self-doping derivatives, and copolymer derivatives.

In addition, the material of organic conductive layer 504 may be a blend of organic conductive material and conventional polymer material, wherein the

conventional polymer is for example polymethyl methacrylate (PMMA), polyvinyl alcohol (PVA), polyethylene oxide (PEO), polyethylene terephthalate (PET), and polyacrylamide (PAA) etc. and the blend ratio of the conventional polymer material is from 1% to 99%.

Referring to Fig. 5C, the organic conductive film on the substrate is patterned for forming regions for displaying pictures and characters as to form patterned organic conductive layer 504a by exposure which, for example, uses a mask 506 with ultraviolet or electron beams to irradiate the regions which do not use for displaying; or uses laser beams under computer controlled to scan on the regions which do not use for displaying. Through above-mentioned method, the regions used for displaying is defined in the organic conductive film.

Referring to Fig. 5D, an organic light emitting layer 508 is formed on the patterned organic conductive layer 504a for example by evaporation or spin-coating. The organic light emitting layer 508 formed is for example 1000 angstroms of poly(2-methoxy, 5-(2'-ethylhexyloxy)-p-phenylene-vinylene (MEH-PPV) or a laminate of N,N'-diphenyl-N,N'-(m-tolyl)benzidine (TPD) of about 500 angstroms in thickness and tri-(8-hydroxyquinoline)aluminum (Alq_3) of about 500 angstroms in thickness. The N,N'-diphenyl-N,N'-(m-tolyl)benzidine is a material for hole transport layer (not shown) which overlying on the surface of the patterned organic conductive layer 504a. Tri-(8-Hydroxyquinoline)aluminum is a material required by organic light emitting layer 508 and overlies on the surface of the hole transport layer as to form an organic light emitting layer 508 with the hole transport material.

Next, referring to Fig. 5E, a metal electrode 510 and a metal protection layer (not shown) are sequentially formed on organic light emitting layer 508 by for example evaporation. The material of metal electrode 510 comprises magnesium (Mg) with a thickness of about 1000 angstroms while that of metal protection layer comprises silver (Ag) with a thickness of about 1000 angstroms.

Accordingly, a structure of this device consisted of transparent conductive layer/patterned organic conductive layer/organic light emitting layer/metal electrode layer/metal protection layer is formed, wherein the organic conductive layer is formed by organic conductive material or a blend of organic conductive material or conventional polymer material while the organic light emitting layer is formed by poly(2-methoxy,5-(2-ethylhexyloxy)-p-phenylene-vinylene layer or a laminate of N,N'-diphenyl-N,N'-(m-tolyl)benzidine and tris-(8-hydroxyquineline)aluminum.

Thus, the material structure of the device may be consisted of said organic conductive material and organic light emitting material, for example, transparent conductive layer/doped polythiophene/poly(2-methoxy, 5-(2'-ethylhexyloxy)-p-phenylene-vinylene/magnesium/silver (TCL/Doped

PT/MEH-PPV/Mg/Ag); transparent conductive layer/doped polyaniline/ poly(2-methoxy, 5-(2'-ethylhexyloxy)-p-phenylene-vinylene/magnesium/silver (TCL/Doped PAn/MEH-PPV/Mg/Ag); transparent conductive layer/doped polythiophene/N,N'-diphenyl-N,N'-(m-tolyl)benzidine/tris-(8-hydroxyquinoline)alum inum/ magnesium/silver (TCL/Doped PT/TPD/Alq_3/Mg/Ag), and transparent conductive layer/doped polyaniline/ N,N'-diphenyl-N,N'-(m-tolyl)benzidine/ tris-(8-hydroxyquinoline)aluminum/ magnesium/silver (TCL/Doped PAn/TPD/Alq_3/Mg/Ag) etc.

The transparent conductive layer 502 and metal electrode 510 are connected with proper operating bias for displaying desired pictures or characters.

From said embodiment of this invention, the present invention provides a manufacturing method for organic electro-luminescent device of static type and presents the following advantages in view of the conventional technologies:

1. The patterning step for transparent conductive layer is omitted such that the required time and costs for placing photoresist, exposure, development, etching and removal of photoresist are eliminated;

2. Either organic light emitting layer or metal electrode does not require patterning step such that the steps of placing photoresist, exposure, development, etching and removal of photoresist are eliminated which would otherwise damage the illumination efficiency of light emitting material and the cleanness of surface. An organic electro-luminescent device of good illumination efficient is achieved;

3. As the unevenness of luminescence of displayed pictures or characters resulted from the gradient distribution of electric field while patterning of transparent conductive layer or metal electrode, the displaying quality is influenced, which is particularly significant in high resolution and low line width display mode. A standard process may be applied to pattern the organic conductive layer of this invention by a ultraviolet or electron beam, the electric field and luminescence of the display regions are even.

4. The manufacturing process of the present invention may control the total exposure amount of the organic conductive film by gradient mask or exposure time for manufacturing a display pattern with luminescence gradation effect and brightness difference.

5. In addition to existing ultraviolet or electron beam process for patterning, a laser beam of computer-controlled may readably and feasibly make any desired pictures or characters and any enclosed pictures or characters which is not displayable by the patterned transparent conductive layer or metal electrode. The process of the present invention presents a simpler and flexible process in comparison with the inkjet process.

6. The method of the present invention is applicable for both a solution coating polymer organic EL device and a small molecule organic EL device by evaporation.

The present invention has been descried using an exemplary preferred embodiment. However, it is to be understood that the scope of the invention is not limited to the disclosed embodiment. On the contrary, it is intended to cover various modifications and similar arrangements. The scope of the present invention should be defined by the following claims.

What is claimed is:

1. A method for manufacturing an organic electro-luminescent device for displaying pictures or characters on a transparent conductive glass substrate, comprising the steps of:

forming an organic conductive layer on said transparent conductive glass substrate;

forming a region for displaying pictures and characters in said organic conductive layer;

forming an organic light emitting layer on said organic conductive layer;

forming a metal electrode on said organic light emitting layer; and

forming said organic electro-luminescent device.

2. The method of claim 1, wherein the step of forming an organic conductive layer includes spin-coating.

3. The method of claim 1, wherein said organic conductive layer is one selected from a group consisting of polyacetylene, polyaniline, polythiophene, poly(phenylene vinylene, poly(paraphenylene), polythiophene vinylene, poly(phenylene sulfide), poly(phenylene oxide), and their derivatives, self-doping derivatives, and copolymer derivatives.

4. The method of claim 1, wherein the material of said organic conductive layer is a blend of an organic conductive material and conventional polymer material.

5. The method of claim 4, wherein the material of said organic conductive layer is one selected from a group consisting of polyacetylene, polyaniline, polythiophene, poly(phenylene vinylene, poly(paraphenylene), polythiophene vinylene, poly(phenylene sulfide), poly(phenylene oxide), and their derivatives, self-doping derivatives, and copolymer derivatives.

6. The method of claim 4, wherein said conventional polymer material is one selected from a group consisting of polymethyl methacrylate, polyvinyl alcohol, polyethylene oxide, polyethylene terephthalate, polyacrylamide.

7. The method of claim 4, wherein the blend ratio of the conventional polymer material is from 1% to 99%.

8. The method of claim 1, wherein the step of forming a region for displaying pictures and characters includes using ultraviolet beams.

9. The method of claim 1, wherein the step of forming a region for displaying pictures and characters includes using electron beams.

10. The method of claim 1, wherein the step of forming a region for displaying pictures and characters includes using laser scanning.

11. The method of claim 1, wherein the step of forming organic light emitting

layer includes spin-coating.

12. The method of claim 1, wherein the step of forming organic light emitting layer includes evaporation.

13. The method of claim 1, wherein the step of forming metal electrode includes evaporation.